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Ultraviolet light amplification within a nanometer-sized layer

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Light amplification in the ultraviolet region is confirmed in mixed rubidium-cesium chloride crystals at room temperature. The probe laser light, which falls on the 275-nm band of Auger-free luminescence (AFL) arising from radiative transition of the Cl $3p$ valence electrons into the Cs $5p$ core holes, is enhanced in intensity when the deep-lying Rb $4p$ core electrons are pumped into the conduction band by undulator radiation from an electron storage ring. The obtained enhancement factor roughly corresponds to an amplification coefficient of 7×10^3 cm^{-1}, which is much higher than those of typical solid-state lasers. It is emphasized that the amplification of AFL occurs in a surface layer as thin as about 20 nm, and that the inverted population between the valence and core bands is realized with any pump power. The present observation provides us with a new possibility of nanolaser fabrication.

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Nanometer-sized lasers (nanolasers) have recently attracted a great deal of attention. From a scientific aspect these lasers present interesting quantum-mechanical phenomena such as the quantum confinement effect or threshold-less laser oscillation. From an application aspect nanolasers are expected to be a key element in realizing an optical computer on a single semiconductor chip or integrated optical fiber communication devices. The main trend of developing such lasers is now directed to semiconductor lasers.[1]

Solid-state laser materials including various types of semiconductors have been intensively investigated, revealing their practical usefulness, e.g., the ease of handling and durability from optical and mechanical points of view. Compared to the development of solid-state lasers emitting wavelengths of light extending from 450 nm in the blue to 10 μm in the infrared, laser-active materials for the ultraviolet region between 180 and 300 nm have been less explored so far. If a solid-state laser, which emits coherent short-wavelength light and has a very small size, becomes a reality, it will open a way for evolving high-speed information processing devices as well as basic sciences.

For the development of short-wavelength nanolasers, we take notice of utilizing Auger-free luminescence.[2,3] A simple quantum theory on atoms, molecules, and monoatomic solid states predicts that the energy separation between the neighboring electronic levels becomes larger for deeper levels as long as they are in a Coulomb-like potential. In such a situation, no intense photoluminescence is emitted even if positive holes are generated in a shallow-lying core state, because the Auger effect inevitably takes place. Interestingly enough, this is not the case in some compound materials where the reverse order of the energy separation can occur for certain levels. When a hole is generated in the outermost-core band of these materials, intrinsic luminescence due to radiative transitions from the valence band to the outermost-core band is observed in a wide spectral range from 160 to 400 nm, without engendering Auger electrons. This specific luminescence is induced by the excitation with photons[2,3] or electron beams,[4] and has been named Auger-free luminescence (AFL).[5] The AFL is characterized by a relatively high yield and a high-temperature stability.[5,6]

Because both the valence and core bands are fully populated in thermal equilibrium, the population inversion between these two bands will easily occur with any intensity of excitation through which some empty (hole) states are created in the lower core band. This means that AFL material is a good candidate for efficient laser-active materials. In contrast to the usual solid-state lasers where active components are impurities doped into host materials, the active component in the present case is AFL material itself. This fact may promise high-density excitation in favor of nanoscale laser action without accompanying any difficulty caused by heavy doping of impurities.

A few experiments on light amplification have recently been carried out in pure and mixed AFL materials.[7,8] They have revealed a sharpening of the emission spectra and a shortening of the luminescence decay time when AFL is retroreflected by a mirror. In this paper we will present more definitive evidence for light amplification of AFL by observing directly an enhancement factor, i.e., an optical gain. The obtained value leads to a surprisingly large amplification coefficient compared to those of the usual solid-state lasers.

In the present experiment we utilized impurity-associated AFL in mixed $Rb_{1-x}Cs_xCl$ ($x=0.18$) crystals. This type of luminescence appears in AFL-free alkali halides (e.g., KCl or RbCl) containing alkali impurity ions with small ionization energy. They form the impurity states in the energy gap between the host valence and outermost-core bands, which provides an additional channel for radiative decay of the core

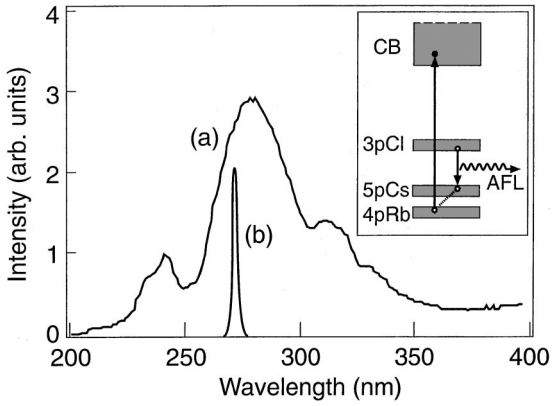

FIG. 1. (a) Luminescence spectrum of a $Rb_{0.82}Cs_{0.18}Cl$ crystal measured at room temperature under the excitation with undulator radiation at 36 eV. No instrumental correction has been made. (b) Spectral profile of the probe laser. Inset: Schematic energy-level diagram showing impurity-associated AFL in mixed rubidium-cesium chloride crystals.

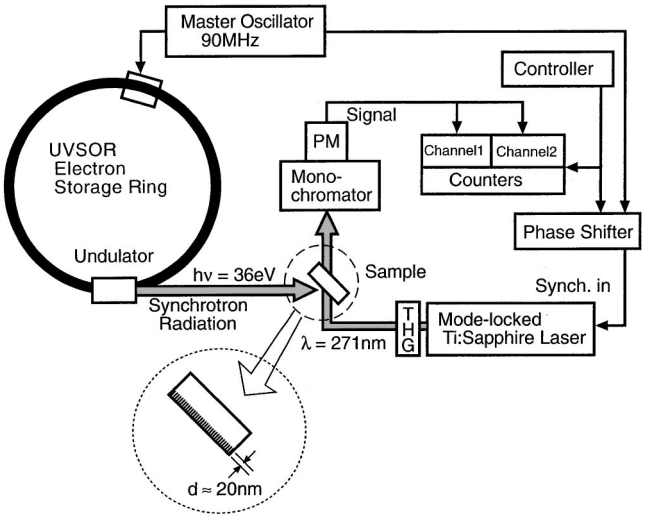

FIG. 2. Block diagram of the experimental setup for light amplification by using pump-and-probe method. Inside the dotted circle is an enlarged view of the sample crystal. PM: microchannel-plate-type photomultiplier, THG: third harmonic generator, and *d*: penetration depth of the undulator light.

holes generated on the impurity states. A schematic energy-level diagram of mixed $Rb_{1-x}Cs_xCl$ is shown in the inset of Fig. 1. The AFL arises from radiative transitions of the host Cl $3p$ valence electrons to the impurity Cs $5p$ core holes, and has a lifetime of about 1 nsec. This AFL spectrum consists of the main band at 275 nm, the high-energy band at 240 nm, and the low-energy tail around 325 nm,[9,10] as presented by a solid curve (a) in Fig. 1. It has been confirmed that the impurity AFL yield in $Rb_{1-x}Cs_xCl$ crystals can significantly exceed the intrinsic AFL yield in pure CsCl, even at small cesium concentration.[10] The observed enhancement of the emission yield has been supposed to be due to the decrease of nonradiative surface losses in the region of the impurity-core absorption, leading to efficient transformation of the incident radiation into photoluminescence.[11] This fact offers therefore fairly attractive conditions for light amplification.

The present experiment was basically a pump-and-probe measurement with synchrotron radiation as pump and laser light as probe. Figure 2 depicts the block diagram of our experimental setup at beam line 3A1 of UVSOR facility, Okazaki. With the use of a permanent magnet undulator, this beam line provides quasimonochromatic light with $\Delta\lambda/\lambda$ 5%, photon flux 10^{15} photons/s mm^2, pulse width 1.5 nsec, and repetition period 11 nsec. The undulator radiation at 36 eV is the most intense (0.1 nJ/pulse) at beam line 3A1.

The highest yield of the 275-nm luminescence occurs when the sample is excited with photons in the region of the Cs $5p$ state located at 15 eV below the bottom of the conduction band, which is five times larger than that excited with 36-eV photons.[10] The luminescence signal excited at 36 eV is, however, greater than that excited at 15 eV, because the photon flux of the undulator radiation at 36 eV is about ten times the one at 15 eV. Hence, the undulator radiation was adjusted to 36 eV in this experiment, corresponding to the transition from the deep-lying Rb $4p$ core band to the conduction band.

A freshly cleaved sample of $Rb_{0.82}Cs_{0.18}Cl$ with 1-mm thickness was mounted on a rotatable holder installed in a vacuum chamber. The undulator radiation was incident at nearly 45° on the sample surface with a spot size of about 1 mm in diameter.

The probe was the third harmonic light (271.5 nm) of a mode-locked Ti:sapphire laser (Coherent Mira 900F). A good spectral coincidence between the probe light and the main 275-nm AFL band is seen in Fig. 1. The laser light of 100 fs in pulse width was synchronized with the undulator radiation by a synchronization system (SynchroLock). The synchronization jitter was found to be less than 100 ps. As shown in Fig. 2, the laser light was incident at a right angle to the undulator light. In order to ensure that the probe intensity is much weaker than the pump intensity, the laser intensity was carefully reduced to becoming comparable to the AFL intensity. The pump and probe beams had nearly the same spot size, and were spatially overlapped on the sample surface. The laser beam transmitted through the pumped domain of the sample was observed using a monochromator (Jobin-Yvon HR320) equipped with a microchannel-plate-type photomultiplier (Hamamatsu R2809U).

The photomultiplier signal was recorded by means of a time-gated photon counting system,[12] employing one-second-wide timing gates. The measurements were performed at room temperature as follows: In the first second of the measurement cycle (phase 1), both the undulator and laser pulses arrived at the sample position simultaneously, and the photon counts were accumulated in counter channel 1 shown in Fig. 2. For the next second (phase 2), a phase shifter inserted between the master oscillator and the synchronization circuit was switched on so that the Ti:sapphire laser pulse was delayed on about 8 ns to the undulator pulse, and the photon counts were accumulated in counter channel 2. The total photon counts for each channel were in the order of 5×10^4 after 700 measurement cycles. The difference in the two counts, channel 1– channel 2, thus yielded an enhancement of the probe caused by the pumping. After com-

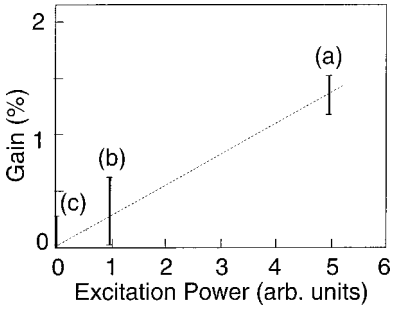

FIG. 3. The measured probe enhancement factor is plotted against the pump power. The pump power in (b) is 1/5 of that in (a), and that in (c) is effectively zero. The dotted line is a guide to the eye.

pensating time dependence of pump power, and averaging the results from five sets of measurements, we obtained the value of 1.5% with dispersion $\pm 0.5\%$ for the probe enhancement, which is well beyond a statistical fluctuation of 0.4%.

In order to further verify the enhancement effect of the probe light, we made the same measurement by changing the pump power under constant laser intensity. In this experiment, (a) the pump power was set at maximum, (b) the pump power was reduced by 1/5 by inserting a metal mesh filter in the beam line, and (c) the spatial overlap of the pump and probe beams was removed, meaning zero excitation at the probed region. The results are shown in Fig. 3. Although there are only three data points, one can see an increase of the enhancement factor with increasing the pump power. From this figure, it is revealed that there is no possibility that the enhancement of the probe signal is due to detection of simple fluorescence induced by the laser light.

The present result of the probe enhancement, as well as its dependence on pump intensity, provides clear evidence for the light amplification of AFL. The undulator radiation at 36 eV creates holes in the deep-lying Rb $4p$ core band. These core holes relax quickly into the impurity Cs $5p$ core band, resulting in the population inversion between the host Cl $3p$ valence band and this impurity core band. It is noteworthy that the population inversion between these two bands is realized without any threshold intensity of excitation. We conclude that the obtained probe enhancement is very likely taken as a light amplification due to stimulated emission in AFL band.

When the enhancement factor ε is small, the amplification coefficient γ is simply approximated as $\gamma=\varepsilon/d$, where d is the penetration depth of the pump light. Taking into account the fact that the penetration depth of the 36-eV light into the host RbCl crystal is about 20 nm,[13,14] we can estimate the amplification coefficient $\gamma=7\times10^3$ cm^{-1} from the enhancement factor 1.5%. This value is about two orders of magnitude larger than those of typical semiconductor lasers; e.g., $\gamma\approx10^2$ cm^{-1} in GaAs-active layer of GaAs-$Al_xGa_{1-x}As$ double heterostructure laser.[15] Such large value of γ promises us that AFL materials have a possibility of very efficient laser action.

Since the enhancement increases with the undulator radiation intensity, we can anticipate laser action if more intense excitation is used to overcome laser cavity loss. Furthermore, it should be noted that no stable coloration and aging effects were seen after prolonged (40-h) irradiation of the samples under 0.9 W/cm^2 of pump irradiance. This fact favorably suggests that the $Rb_{1-x}Cs_xCl$ mixed crystals indeed possess a promising potentiality as a realistic laser medium. When a laser medium is pumped with a fixed repetition period as in our experiment, mode-locking operation of the laser is expected if its cavity round trip time matches with the repetition period of the pump. In this case, the output laser pulse width as short as 10 fs will be realized by considering the observed spectral width ($\sim$30 nm) of the main 275-nm band in the present material.

For a higher optical gain, the excitation of a thick layer ($\gg$20 nm) is supposed to be preferable. One approach to this direction is to use x rays as a pump source. Another approach is to apply a nano lithography technique to our system. It would be possible to fabricate a one-dimensional periodic array of $Rb_{1-x}Cs_xCl$ crystal with 20-nm thickness on an appropriate substrate. When the outermost-core band (Cs $5p$ or Rb $4p$) of each nanocrystal is pumped by photons or electron beams, amplified AFL is expected to appear along the array.

In conclusion, we have observed the light amplification of AFL in mixed rubidium-cesium chloride crystals by pump-and-probe experiment. The laser-active region of the present material is a surface layer as thin as 20 nm. This type of light amplification will open up the possibilities for development of nanometer-sized solid-state lasers operating in the ultraviolet region.

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